

Supplementary Information for "DFT investigation of Na-ion interaction with Janus 1T-HfSTe monolayer for sodium-ion battery anodes"

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TABLE S1. The convergence test of the total energy of the 3×3 supercell with respect to the plane-wave cutoff energy using a $6 \times 6 \times 1$ K-points grid. The small energy variation around the selected cutoff energy of 550 eV indicates that this value is sufficiently reliable for subsequent calculations.

Cutoff energy (eV)	Total energy (eV)
250	-193.88754422
300	-194.00814463
350	-194.05522178
400	-194.05358086
450	-194.08822627
500	-194.08502819
520	-194.08778180
540	-194.08596357
560	-194.07840107
580	-194.08110910

TABLE S2. The convergence test of the total energy of the 3×3 supercell with respect to the K-points grid. The small energy difference between the $5 \times 5 \times 1$ and $6 \times 6 \times 1$ grids, approximately 0.495 meV/atom, indicates that the $5 \times 5 \times 1$ grid is sufficiently reliable for subsequent calculations.

K-points grid	Total energy (eV)	ΔE per atom (meV/atom)
$2 \times 2 \times 1$	-194.13497787	3.011
$3 \times 3 \times 1$	-194.06185039	0.303
$4 \times 4 \times 1$	-194.04681040	0.254
$5 \times 5 \times 1$	-194.06704254	0.495
$6 \times 6 \times 1$	-194.05366832	0.000

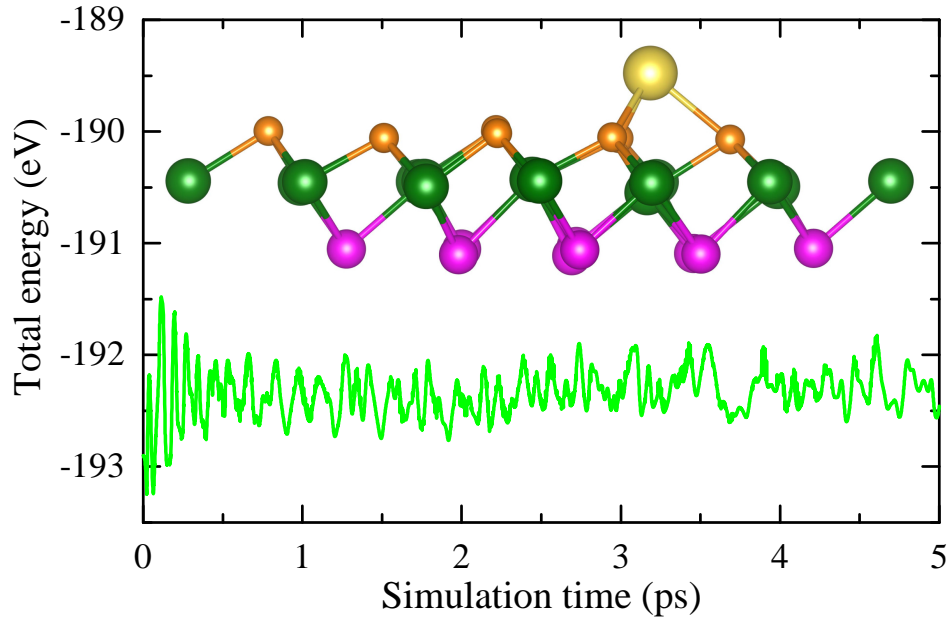


FIG. S1. Ab initio molecular dynamics (AIMD) results for the configuration with the first Na ion adsorbed at the most stable H_1^T site on the HfSTe monolayer. The simulations were performed using a $3 \times 3 \times 1$ supercell in the NVT ensemble with the Nosé–Hoover thermostat at 300 K for 5 ps with a time step of 1 fs. The total-energy evolution and the final structure after 5 ps indicate that the adsorbed Na atom remains stably bound and the HfSTe framework preserves its structural integrity without obvious structural collapse.

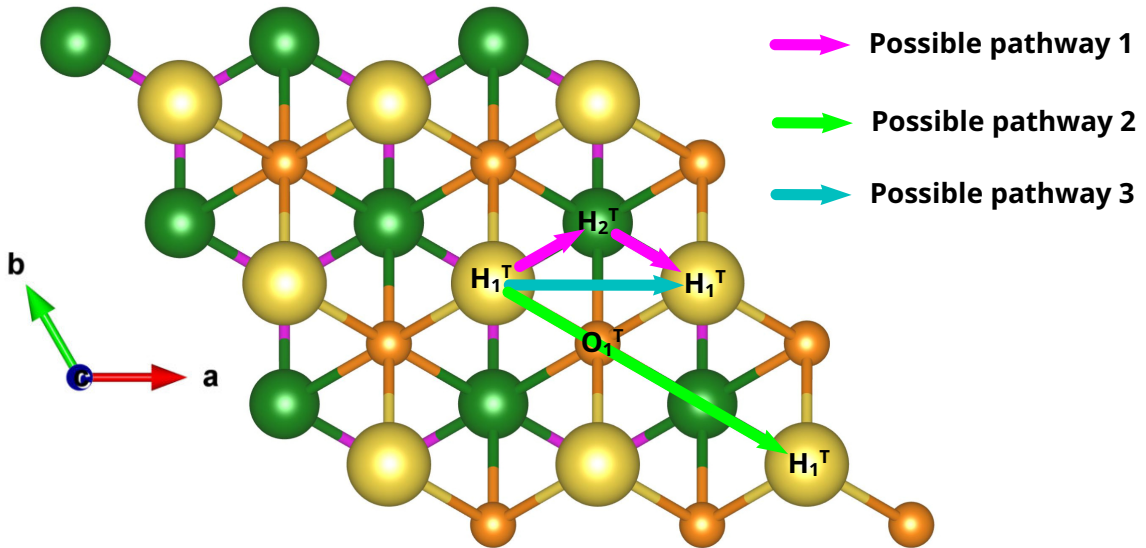


FIG. S2. Three possible Na-ion migration pathways on the S-terminated surface of the Janus HfSTe monolayer: pathway 1 ($H_1^T \rightarrow H_2^T \rightarrow H_1^T$, magenta arrow), pathway 2 (direct $H_1^T \rightarrow H_1^T$ at $\sim 30^\circ$ to the a -axis, green arrow), and pathway 3 (direct $H_1^T \rightarrow H_1^T$ parallel to the a -axis, cyan arrow). Pathway 3 relaxes onto the trajectory of pathway 1 during CI-NEB optimization and is not treated as an independent route. Orange, dark green, and gold spheres represent S, Hf, and Na atoms, respectively.

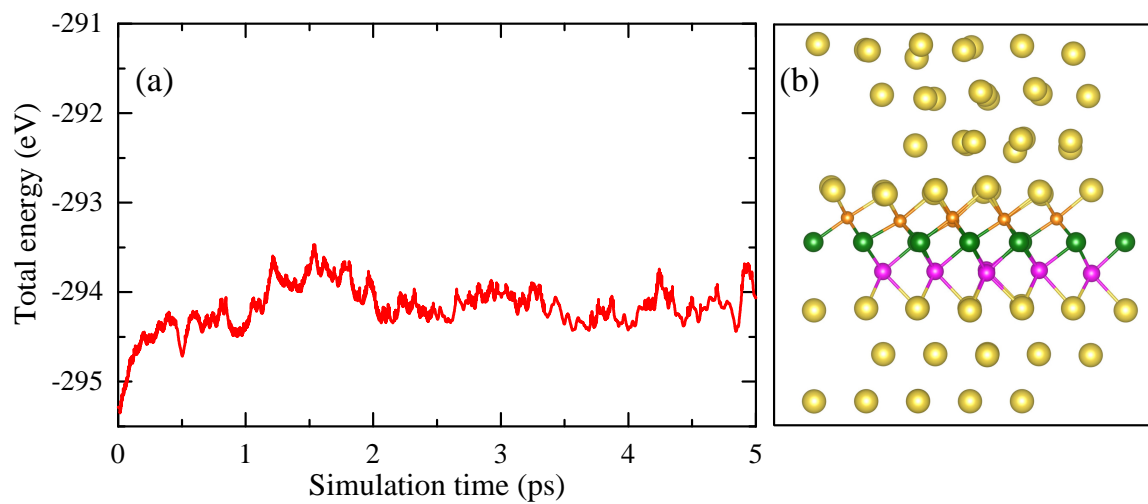


FIG. S3. AIMD results for the fully sodiated Na_7HfSTe configuration. The simulations were performed in the NVT ensemble with the Nosé–Hoover thermostat at 300 K for 5 ps with a time step of 1 fs. The total-energy evolution and the final structure after 5 ps confirm the thermal stability of Na_7HfSTe without obvious structural collapse.

OPTIMIZED STRUCTURE OF HFSTE MONOLAYER

Hf S Te

0.9800000000000000

3.8545137563775360 0.0000000000000000 -0.0000000000000000

-1.9272568781887680 3.3381068322593235 0.0000000000000000

0.0000000000000000 -0.0000000000000000 19.0184079539224697

Hf S Te

1 1 1

Selective dynamics

Direct

0.0000000000000000 0.0000000000000000 0.5108643409534956

0.6666666999999979 0.333333300000021 0.5805739041544518

0.333333300000021 0.6666666999999979 0.4085609148920471